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Antiferromagnetic instability of heavy-fermion alloys with conduction band impurities

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Abstract. The antiferromagnetic instability of heavy-fermion alloys with conduction band impurities is examined in the single-site coherent-potential approximation with the strong correlation treated by the slave-boson technique of Kotliar and Ruckenstein. The variation of the paramagnetic–antiferromagnetic phase boundary with the impurity concentration is studied in the case of isoelectronic doping. Our results show that the isoelectronic doping favours the formation of antiferromagnetism if the impurities increase the cell volume, thus suppressing the c–f mixing, which is in agreement with experimental observations in $Ce(Cu_{1-x}Ag_x)_6$ and $Ce(Cu_{1-x}Au_x)_6$. The obtained phase diagram can also be used to explain the effect of non-isoelectronic substitutions of Al for Cu in $CeCu_6$.

1. Introduction

The weak instability against magnetism is an important topic in research on normal heavy-fermion (HF) systems, and has received a large amount of attention in recent years. Many pieces of evidence show that HF systems totter on the very edge of magnetism. Neutron scattering experiments detected that antiferromagnetic correlations in $CeCu_6$ develop below 1 K [1]. For $CeCu_2Si_2$, μSR results at $B = 0$ T are consistent with the onset of long-range antiferromagnetism at $T_N = 0.8$ K [2]. Many normal HF compounds have been reported to emerge with tiny ordered moments [3–5].

Controlled alloying is regarded as an effective way of studying the properties of normal HF compounds, and in particular the proximity to magnetism. It has been indicated that the ground states of these compounds can be modified by substitutions on either f element sites or non-f metal sites [6]. In $CeCu_6$, substitutions of Au or Ag on Cu sites (so-called isoelectronic doping (IED)), have been found to produce magnetic order [7–9], whereas Al substitutions for Cu give rise to no magnetic order [6]. Lees and co-workers suggested that these phenomena, particularly in the case of IED, may be attributed to the lattice spacing variation induced by the alloying [6]. This is reasonable because the HF systems have such an anomalously large electronic Gruneisen constant [10] that the competition between spin compensation and magnetic order should be sensitive to the lattice spacing or cell volume.

In order to shed light on the complicated and multiplex behaviour of the HF magnetism revealed in experiments, much theoretical effort has been focused on slave-boson based techniques, which were introduced by Barnes and extended by others [11–14]. Dorin and Schlottmann studied the magnetic instability of Kondo insulators in the symmetric Anderson lattice [15]. Reynolds and co-workers calculated the magnetism as a function of applied magnetic field in the ferromagnetic state of the periodic Anderson model (PAM) [16]. Furthermore, treating the slave bosons as spin-carrying bosons, Möller

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and Wölfle studied the spiral magnetic order of the symmetric PAM [17]. Chen and Li investigated the antiferromagnetic instability of the Anderson lattice model, and the paramagnetic–antiferromagnetic phase boundary was obtained [18]. However, the influence of conduction-band (CB) impurities on the phase boundary have not been discussed. In the present paper, we intend to provide a systematic study of the alloying effects on the antiferromagnetic instability under the Kotliar and Ruckenstein (KR) slave-boson scheme by adopting the single-site coherent-potential approximation (CPA) method to discuss the influence of CB impurities on the phase boundary, so that its dependence on the impurity concentration can be calculated.

The paper is organized as follows. In section 2, the KR slave-boson mean-field (SBMF) Hamiltonian of the HF alloys is presented. In section 3, the effective medium and the mean-field parameters are determined self-consistently. In section 4, we calculate the paramagnetic–antiferromagnetic phase boundary of the HF alloys, and the effect on magnetic instability of CB impurities is obtained. In section 5 we summarize our theoretical investigation.

2. The Hamiltonian of HF alloys with CB impurities

The PAM is considered as the essential description of the HF system [19]. We start with this model in order to discuss the effects of CB impurities. Since the main effect of the impurities is to bring about a change in the cell volume of the impurity site, thus giving rise to a change in the c–f mixing strength, the Hamiltonian of HF alloys can be written as

$$H = t\sum_{l,r,\sigma} c^{+}_{l+r,\sigma}c_{l,\sigma} + \sum_{l,\sigma}(V+\xi_l\Delta V)(c^{+}_{l,\sigma}f_{l,\sigma}z_{l,\sigma} + z^{+}_{l,\sigma}f^{+}_{l,\sigma}c_{l,\sigma}) + \sum_{l,\sigma}E_0 f^{+}_{l,\sigma}f_{l,\sigma} + U\sum_{l} d^{+}_{l}d_{l} + \sum_{l}\lambda^{(1)}_{l}(e^{+}_{l}e_{l} + d^{+}_{l}d_{l} + \sum_{\sigma}p^{+}_{l,\sigma}p_{l,\sigma} - 1) + \sum_{l,\sigma}\lambda^{(2)}_{l,\sigma}(f^{+}_{l,\sigma}f_{l,\sigma} - p^{+}_{l,\sigma}p_{l,\sigma} - d^{+}_{l}d_{l}) \tag{1}$$

where $c^{+}_{l,\sigma}(f_{l,\sigma})$ creates (annihilates) a conduction (localized) electron in the Wannier state at site l with spin σ; r and t denote respectively the relative displacement and the c-electron hopping integral between the nearest-neighbour sites, ξ_l is the random variable with $\xi_l = 0$ for a site l with no impurities and $\xi_l = 1$ for a CB-impurity-occupied site l, where the c–f mixing strength V changes by ΔV. In Hamiltonian (1), we have introduced the KR slave bosons for each f site. They satisfy the constraints

$$e^{+}_{l}e_{l} + d^{+}_{l}d_{l} + \sum_{\sigma}p^{+}_{l,\sigma}p_{l,\sigma} = 1 \tag{2}$$

$$f^{+}_{l,\sigma}f_{l,\sigma} = d^{+}_{l}d_{l} + p^{+}_{l,\sigma}p_{l,\sigma}, \sigma = \pm 1 \tag{3}$$

which are incorporated via Lagrange multipliers $\lambda^{(1)}_{l}$ and $\lambda^{(2)}_{l,\sigma}$ respectively. The operator $z_{l,\sigma}$ with the definition

$$z_{l,\sigma} = (1 - d^{+}_{l}d_{l} - p^{+}_{l,\sigma}p_{l,\sigma})^{-1/2}(e^{+}_{l}p_{l,\sigma} + p^{+}_{l,-\sigma}d_{l})(1 - e^{+}_{l}e_{l} - p^{+}_{l,-\sigma}p_{l,-\sigma})^{-1/2} \tag{4}$$

is used not only to keep the matrix elements of $f^{+}_{l,\sigma}$ and $f_{l,\sigma}$ invariant in the combined fermion–boson Hilbert space, but also, at the saddle-point level, to ensure the correct

non-interacting limit and the reproduction of the Gutzwiller approximation in the strong-correlation limit [20].

In order to study the antiferromagnetic instability, we introduce an infinitesimal staggered magnetic field

$$h_l = h\mathrm{e}^{\mathrm{i}Q\cdot l} \tag{5}$$

which is applied on electrons, hence adding a Zeeman term $\sigma E_1 \mathrm{e}^{\mathrm{i}Q\cdot l}$ to both the c- and f-electron energies. Here $E_1 = -\mu_B h$ and Q is a wavevector that satisfies the staggered condition $\mathrm{e}^{\mathrm{i}Q\cdot r} = -1$. In the paramagnetic state, the applied field will produce a staggered polarization of c and f electrons. Within the mean-field approximation we take

$$p^+_{l,\sigma} = p_{l,\sigma} = p_0 + \sigma p_1 \mathrm{e}^{\mathrm{i}Q\cdot l} \tag{6}$$

$$\lambda^{(2)}_{l,\sigma} = \lambda^{(2)}_0 + \sigma\lambda^{(2)}_1 \mathrm{e}^{\mathrm{i}Q\cdot l} \tag{7}$$

$$e^+_l = e_l = e \tag{8}$$

$$d^+_l = d_l = d \tag{9}$$

$$\lambda^{(1)}_l = \lambda^{(1)} \tag{10}$$

where p_0, $\lambda^{(2)}_0$ and $\lambda^{(1)}$ can be determined in the absence of h_l, and p_1, $\lambda^{(2)}_1$ are the linear responses of h_l. Correspondingly, the renormalization parameter $z_{l,\sigma}$ is also of staggered form:

$$z_{l,\sigma} = z_0 + \sigma z_1 \mathrm{e}^{\mathrm{i}Q\cdot l}. \tag{11}$$

To simplify the CPA formalism in section 3, we introduce two interpenetrating sublattices a and b whose site vectors i and j satisfy $\mathrm{e}^{\mathrm{i}Q\cdot i} = 1$ and $\mathrm{e}^{\mathrm{i}Q\cdot j} = -1$ respectively. Since the number of cells of the sublattice is half that of the original lattice sites N, the magnetic Brillouin zone (MBZ) is half the original BZ. Using Fourier transformations in the sublattices

$$C^a_{i,\sigma} = (\tfrac{1}{2}N)^{-1/2} \sum_k \mathrm{e}^{\mathrm{i}k\cdot i} C^a_{k,\sigma} \tag{12}$$

$$C^b_{j,\sigma} = (\tfrac{1}{2}N)^{-1/2} \sum_k \mathrm{e}^{\mathrm{i}k\cdot j} C^b_{k,\sigma} \tag{13}$$

with the two-component fields in sublattice $a(b)$, $C^{a(b)}_{i(j),\sigma} \equiv (c^{a(b)}_{i(j),\sigma}, f^{a(b)}_{i(j),\sigma})^{\mathrm{T}}$ and $C^{a(b)}_{k,\sigma} \equiv (c^{a(b)}_{k,\sigma}, f^{a(b)}_{k,\sigma})^{\mathrm{T}}$, we get the mean-field Hamiltonian in the paramagnetic state:

$$H = \sum_{k,\sigma} (C^{a+}_{k,\sigma}, C^{b+}_{k,\sigma}) \mathcal{E}_{k,\sigma} \begin{pmatrix} C^a_{k,\sigma} \\ C^b_{k,\sigma} \end{pmatrix} + \sum_{l,\sigma} \xi_l C^+_{l,\sigma} \Delta\mathcal{V}_{l,\sigma} C_{l,\sigma} + NUd^2$$
$$+ N\lambda^{(1)}\left(e^2 + d^2 + 2p_0^2 + 2p_1^2 - 1\right) - 2N\left(\lambda^{(2)}_0 d^2 + \lambda^{(2)}_0 p_0^2 + \lambda^{(2)}_0 p_1^2 + 2\lambda^{(2)}_1 p_0 p_1\right). \tag{14}$$

Here, and hereinafter, the summations over k are restricted within the MBZ; $C_{l,\sigma}$, $\Delta\mathcal{V}_{l,\sigma}$ and $\mathcal{E}_{k,\sigma}$ are defined as follows:

$$C_{l,\sigma} \equiv (c_{l,\sigma}, f_{l,\sigma})^{\mathrm{T}}$$

$$\Delta\mathcal{V}_{l,\sigma} = \Delta\mathcal{V}_0 + \sigma\Delta\mathcal{V}_1 e^{iQ\cdot l} \equiv \begin{pmatrix} 0 & z_0\Delta V \\ z_0\Delta V & 0 \end{pmatrix} + \sigma \begin{pmatrix} 0 & z_1\Delta V \\ z_1\Delta V & 0 \end{pmatrix} e^{iQ\cdot l} \tag{15}$$

$$\mathcal{E}_{k,\sigma} \equiv \begin{pmatrix} 0 & z_0V & \varepsilon_k & 0 \\ z_0V & E_0+\lambda_0^{(2)} & 0 & 0 \\ \varepsilon_k & 0 & 0 & z_0V \\ 0 & 0 & z_0V & E_0+\lambda_0^{(2)} \end{pmatrix} + \sigma \begin{pmatrix} E_1 & z_1V & 0 & 0 \\ z_1V & E_1+\lambda_1^{(2)} & 0 & 0 \\ 0 & 0 & -E_1 & -z_1V \\ 0 & 0 & -z_1V & -E_1-\lambda_1^{(2)} \end{pmatrix} \tag{16}$$

with the dispersion of the conduction band

$$\varepsilon_k = t\sum_r e^{\pm ik\cdot r}. \tag{17}$$

For simplicity, the density of states (DOS) of the unperturbed c-electrons is assumed to be constant with a bandwidth of $2D$. We would like to caution the reader that the appearance of the c-electron energy ε_k as off-diagonal elements is due to the c-electron hopping integral between the nearest-neighbour sites belonging to different sublattices.

Because of the random variable ξ_l, the Hamiltonian (14) cannot be diagonalized by the standard procedure. We shall adopt the single-site CPA to solve this disordered system self-consistently.

3. Effective medium

In the framework of the single-site CPA, the HF disordered system can be imitated by an effective medium in which, on the average, the scattering by the impurities vanishes at each site [21–23]. Considering the l-dependence of the coherent potential due to the staggered form of h_l, we write universally the effective medium Hamiltonian as

$$\tilde{H} = \sum_{k,\sigma}(C_{k,\sigma}^{a+}, C_{k,\sigma}^{b+})\mathcal{E}_{k,\sigma}\begin{pmatrix} C_{k,\sigma}^{a} \\ C_{k,\sigma}^{b} \end{pmatrix} + \sum_{l,\sigma} C_{l,\sigma}^{+}\mathcal{S}_{l,\sigma}(\omega)C_{l,\sigma} + N\bar{U}d^2 + N\lambda^{(1)}\left(e^2+d^2+2p_0^2+2p_1^2-1\right) - 2N\left(\lambda_0^{(2)}d^2+\lambda_0^{(2)}p_0^2+\lambda_0^{(2)}p_1^2+2\lambda_1^{(2)}p_0p_1\right) \tag{18}$$

where the ω-dependent coherent potential $\mathcal{S}_{l,\sigma}$ must be calculated self-consistently from the following single-site CPA equation [21]:

$$(1-c_I)\mathcal{T}_{l,\sigma}(0) + c_I\mathcal{T}_{l,\sigma}(1) = 0. \tag{19}$$

Here $c_I \equiv 1/N\sum_l \xi_l$ is the concentration of CB impurities and $\mathcal{T}_{l,\sigma}$ is the scattering t-matrix:

$$\mathcal{T}_{l,\sigma}(\xi_l) = (\xi_l\Delta\mathcal{V}_{l,\sigma} - \mathcal{S}_{l,\sigma})[1 - \mathcal{F}_{l,\sigma}(\xi_l\Delta\mathcal{V}_{l,\sigma} - \mathcal{S}_{l,\sigma})]^{-1} \tag{20}$$

with the site Green function (GF) $\mathcal{F}_{l,\sigma}(\omega) \equiv \langle\langle C_{l,\sigma}|C_{l,\sigma}^{+}\rangle\rangle_\omega$ corresponding to the effective Hamiltonian $\tilde{H}$.

Apparently, the scattering matrix $\mathcal{T}_{l,\sigma}(\xi_l)$ involves the h-ordered term. In order for us to treat them in the limit $h \to 0$, the specific l-dependence of $\mathcal{S}_{l,\sigma}$ must be revealed. From (18)–(20), it is easily found that $\mathcal{S}_{l,\sigma}$ depends on $\mathcal{E}_{k,\sigma}$ and $\Delta\mathcal{V}_{l,\sigma}$, and thus is a function of the f-electron energy lever $E_{l,\sigma} = E_0 + \sigma E_1 e^{iQ\cdot l}$ and the SBMF parameters $z_{l,\sigma}$ and $\lambda^{(2)}_{l,\sigma}$. We expand it formally near E_0, z_0 and λ_0, then get the same form as $E_{l,\sigma}$, $z_{l,\sigma}$ and $\lambda_{l,\sigma}$:

$$\mathcal{S}_{l,\sigma} = \mathcal{S}_0 + \sigma \mathcal{S}_1 e^{iQ\cdot l} \tag{21}$$

where

$$\begin{aligned}\mathcal{S}_1 &= \frac{\partial \mathcal{S}_{l,\sigma}(E_0, z_0, \lambda_0)}{\partial E_{l,\sigma}} E_1 + \frac{\partial \mathcal{S}_{l,\sigma}(E_0, z_0, \lambda_0)}{\partial z_{l,\sigma}} z_1 + \frac{\partial \mathcal{S}_{l,\sigma}(E_0, z_0, \lambda_0)}{\partial \lambda^{(2)}_{l,\sigma}} \lambda^{(2)}_1 \\ &= \frac{\partial \mathcal{S}_1}{\partial E_1} E_1 + \frac{\partial \mathcal{S}_1}{\partial z_1}\frac{\partial z_1}{\partial p_1} p_1 + \frac{\partial \mathcal{S}_1}{\partial \lambda^{(2)}_1}\lambda^{(2)}_1\end{aligned} \tag{22}$$

is of order h. Evidently, the staggered coherent potential (21) processes the periodicity in either sublattice a or b.

Furthermore, the site GF $\mathcal{F}_{l,\sigma}$ can be discussed. With the 4×4 matrix expression in the Bloch representation of $\mathcal{S}_{l,\sigma}$

$$\mathcal{S}_\sigma = \begin{pmatrix} \mathcal{S}_0 + \sigma \mathcal{S}_1 & 0 \\ 0 & \mathcal{S}_0 - \sigma \mathcal{S}_1 \end{pmatrix} \tag{23}$$

the effective medium Hamiltonian is

$$\begin{aligned}\tilde{H} = \sum_{k,\sigma} (C^{a+}_{k,\sigma}, C^{b+}_{k,\sigma})(\mathcal{E}_{k,\sigma} + \mathcal{S}_\sigma) \begin{pmatrix} C^a_{k,\sigma} \\ C^b_{k,\sigma} \end{pmatrix} + NUd^2 \\ + N\lambda^{(1)}\left(e^2 + d^2 + 2p_0^2 + 2p_1^2 - 1\right) - 2N\left(\lambda^{(2)}_0 d^2 + \lambda^{(2)}_0 p_0^2 + \lambda^{(2)}_0 p_1^2 + 2\lambda^{(2)}_1 p_0 p_1\right)\end{aligned} \tag{24}$$

from which the medium GF with momentum k

$$\mathcal{G}_{k,\sigma}(\omega) = (\omega - \mathcal{E}_{k,\sigma} - \mathcal{S}_\sigma)^{-1} \tag{25}$$

is derived. With the dispersion (17), the site GF of the effective medium

$$\mathcal{F}_\sigma(\omega) = (\tfrac{1}{2}N)^{-1} \sum_k \mathcal{G}_{k,\sigma}(\omega) \tag{26}$$

is found to be of quasi-diagonal form:

$$\begin{aligned}\mathcal{F}^{aa}_\sigma &= \mathcal{F}_0 + \sigma \mathcal{F}_1 \\ \mathcal{F}^{bb}_\sigma &= \mathcal{F}_0 - \sigma \mathcal{F}_1 \\ \mathcal{F}^{ab}_\sigma &= \mathcal{F}^{ba}_\sigma = 0.\end{aligned} \tag{27}$$

The result can be generalized by a staggered-formed site GF:

$$\mathcal{F}_{l,\sigma} = \mathcal{F}_0 + \sigma \mathcal{F}_1 e^{iQ\cdot l}. \tag{28}$$

Because $\mathcal{F}_1$ is of order h, it also has an expanded form:

$$\mathcal{F}_1 = \frac{\partial \mathcal{F}_1}{\partial E_1} E_1 + \frac{\partial \mathcal{F}_1}{\partial z_1}\frac{\partial z_1}{\partial p_1} p_1 + \frac{\partial \mathcal{F}_1}{\partial \lambda_1^{(2)}}\lambda_1^{(2)}. \tag{29}$$

In view of the above results, the scattering matrix $\mathcal{T}_{l,\sigma}(\xi_l)$ can also be divided into two terms: a zeroth-order term irrelevant to the staggered field h_l

$$\mathcal{T}_0(\xi_l) = (\xi_l \Delta \mathcal{V}_0 - \mathcal{S}_0)[1 - \mathcal{F}_0(\xi_l \Delta \mathcal{V}_0 - \mathcal{S}_0)]^{-1} \tag{30}$$

and an infinitesimal term responding linearly to h_l

$$\mathcal{T}_1(\xi_l) = [1 - (\xi_l \Delta \mathcal{V}_0 - \mathcal{S}_0)\mathcal{F}_0]^{-1}(\xi_l \Delta \mathcal{V}_1 - \mathcal{S}_1)[1 - \mathcal{F}_0(\xi_l \Delta \mathcal{V}_0 - \mathcal{S}_0)]^{-1} + \mathcal{T}_0(\xi_l)\mathcal{F}_1\mathcal{T}_0(\xi_l). \tag{31}$$

Correspondingly, the single-site CPA (19) is rewritten as two simultaneous equations:

$$(1 - c_1)\mathcal{T}_0(0) + c_1\mathcal{T}_0(1) = 0 \tag{32}$$

$$(1 - c_1)\mathcal{T}_1(0) + c_1\mathcal{T}_1(1) = 0. \tag{33}$$

Noting that the partial derivatives with respect to E_1, z_1 and $\lambda_1^{(2)}$ of infinitesimal parameters such as $\Delta\mathcal{V}_1$, $\mathcal{S}_1$ and $\mathcal{F}_1$ are determinable, we have to evaluate $\partial\mathcal{S}_1/\partial E_1$, $\partial\mathcal{S}_1/\partial z_1$ and $\partial\mathcal{S}_1/\partial\lambda_1^{(2)}$ self-consistently by solving the three independent equations

$$(1 - c_1)\frac{\partial \mathcal{T}_1(0)}{\partial E_1} + c_1\frac{\partial \mathcal{T}_1(1)}{\partial E_1} = 0 \tag{34}$$

$$(1 - c_1)\frac{\partial \mathcal{T}_1(0)}{\partial z_1} + c_1\frac{\partial \mathcal{T}_1(1)}{\partial z_1} = 0 \tag{35}$$

$$(1 - c_1)\frac{\partial \mathcal{T}_1(0)}{\partial \lambda_1^{(2)}} + c_1\frac{\partial \mathcal{T}_1(1)}{\partial \lambda_1^{(2)}} = 0 \tag{36}$$

which are the linear expansion forms of (33) and which satisfy it in all cases.

The mean-field parameters as shown in (6)–(10) will be determined by the minimum condition of the free energy combined with the CPA equations, as is embodied in the following saddle-point equations:

$$\sum_\sigma \langle f_{l,\sigma}^+ f_{l,\sigma}\rangle = 2(p_0^2 + d^2) \tag{37}$$

$$(V + c_1\Delta V)\frac{\partial z_0}{\partial e}\sum_\sigma \langle c_{l,\sigma}^+ f_{l,\sigma} + f_{l,\sigma}^+ c_{l,\sigma}\rangle + 2\lambda^{(1)} e = 0 \tag{38}$$

$$(V + c_1\Delta V)\frac{\partial z_0}{\partial p_0}\sum_\sigma \langle c_{l,\sigma}^+ f_{l,\sigma} + f_{l,\sigma}^+ c_{l,\sigma}\rangle + 4(\lambda^{(1)} - \lambda_0^{(2)})p_0 = 0 \tag{39}$$

$$(V + c_1\Delta V)\frac{\partial z_0}{\partial d}\sum_\sigma \langle c_{l,\sigma}^+ f_{l,\sigma} + f_{l,\sigma}^+ c_{l,\sigma}\rangle + 2(U + \lambda^{(1)} - 2\lambda_0^{(2)})d = 0 \tag{40}$$

$$(V + c_1\Delta V)\frac{\partial z_0}{\partial p_1}\sum_\sigma \langle c_{l,\sigma}^+ f_{l,\sigma} + f_{l,\sigma}^+ c_{l,\sigma}\rangle + (V + c_1\Delta V)\frac{\partial z_1}{\partial p_1}\sum_\sigma \sigma e^{iQ\cdot l}\langle c_{l,\sigma}^+ f_{l,\sigma} + f_{l,\sigma}^+ c_{l,\sigma}\rangle$$
$$+ 4(\lambda^{(1)} p_1 - \lambda_0^{(2)} p_1 - \lambda_1^{(2)} p_0) = 0 \tag{41}$$

$$\sum_\sigma \sigma e^{iQ\cdot l}\langle f_{l,\sigma}^+ f_{l,\sigma}\rangle = 4 p_0 p_1 \tag{42}$$

$$e^2 + d^2 + 2p_0^2 = 1. \tag{43}$$

The chemical potential μ is determined by the total number of electrons:

$$\sum_{\sigma}\langle c_{l,\sigma}^{+}c_{l,\sigma}+f_{l,\sigma}^{+}f_{l,\sigma}\rangle=n_{\mathrm{c}}+1 \tag{44}$$

where n_{c} is the unperturbed conduction electron number per site.

4. Antiferromagnetic instability of HF alloys

The localized susceptibility of HF alloys with CB impurities in the paramagnetic state can be written as

$$\chi_Q=\lim_{h\to 0}\sum_{\sigma}\frac{\sigma\mu_{\mathrm{B}}}{h}\mathrm{e}^{\mathrm{i}Q\cdot l}\langle f_{l,\sigma}^{+}f_{l,\sigma}\rangle=\lim_{h\to 0}\frac{4p_0p_1\mu_{\mathrm{B}}}{h}. \tag{45}$$

As the criterion of antiferromagnetic instability, the divergence of χ_Q means that there exists a non-zero solution of p_1 at zero external field, i.e. $E_1(\equiv -\mu_{\mathrm{B}}h)=0$, in (41) and (42). After some algebraic treatment of the first-order saddle-point equations (41) and (42) by expanding $\mathcal{F}_1$ as (29), one can get a system of homogeneous linear equations of p_1 and $\lambda_1^{(2)}$ when taking $E_1=0$:

$$\begin{pmatrix}\alpha-2p_0 & \beta\\ \gamma+\lambda^{(1)}-\lambda_0^{(2)} & \delta-p_0\end{pmatrix}\begin{pmatrix}p_1\\ \lambda_1^{(2)}\end{pmatrix}=0 \tag{46}$$

where

$$\alpha=-\frac{1}{\pi}\frac{\partial z_1}{\partial p_1}\int_{-\infty}^{\infty}\mathrm{d}\omega f(\omega-\mu)\mathrm{Im}\frac{\partial F_{\mathrm{ff1}}(\omega+\mathrm{i}0^{+})}{\partial z_1} \tag{47}$$

$$\beta=-\frac{1}{\pi}\int_{-\infty}^{\infty}\mathrm{d}\omega f(\omega-\mu)\mathrm{Im}\frac{\partial F_{\mathrm{ff1}}(\omega+\mathrm{i}0^{+})}{\partial \lambda_1^{(2)}} \tag{48}$$

$$\gamma=-\frac{V+c_{\mathrm{I}}\Delta V}{\pi}\left[\frac{\partial^2 z_0}{\partial p_1^2}\int_{-\infty}^{\infty}\mathrm{d}\omega f(\omega-\mu)\mathrm{Im}F_{\mathrm{cf0}}(\omega+\mathrm{i}0^{+})+\left(\frac{\partial z_1}{\partial p_1}\right)^2\int_{-\infty}^{\infty}\mathrm{d}\omega f(\omega-\mu)\mathrm{Im}\frac{\partial F_{\mathrm{cf1}}(\omega+\mathrm{i}0^{+})}{\partial z_1}\right] \tag{49}$$

$$\delta=-\frac{V+c_{\mathrm{I}}\Delta V}{\pi}\frac{\partial z_1}{\partial p_1}\int_{-\infty}^{\infty}\mathrm{d}\omega f(\omega-\mu)\mathrm{Im}\frac{\partial F_{\mathrm{cf1}}(\omega+\mathrm{i}0^{+})}{\partial \lambda_1^{(2)}} \tag{50}$$

with Fermi function $f(\omega-\mu)$ and F_{cf0} and F_{cf1}, F_{ff1} for the matrix elements of the site GF $\mathcal{F}_0$ and $\mathcal{F}_1$ respectively. The existence of non-zero solutions demands that the coefficient determinant of (46) should be equal to zero:

$$(\alpha-2p_0)(\delta-p_0)-\beta(\gamma+\lambda^{(1)}-\lambda_0^{(2)})=0. \tag{51}$$

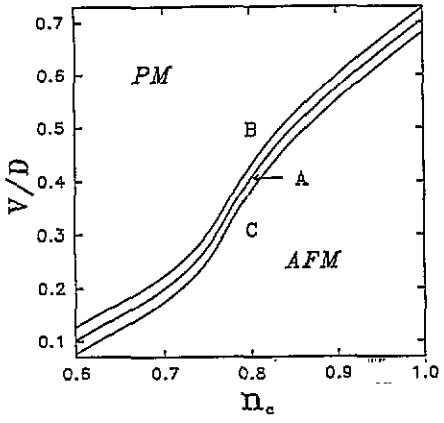

Figure 1. The paramagnetic–antiferromagnetic phase diagram in the n_c–V/D plane; phase boundary A: $c_I = 0$; phase boundary B: $c_I = 0.1$, $\Delta V/D = -0.2$; phase boundary C: $c_I = 0.1$, $\Delta V/D = 0.2$.

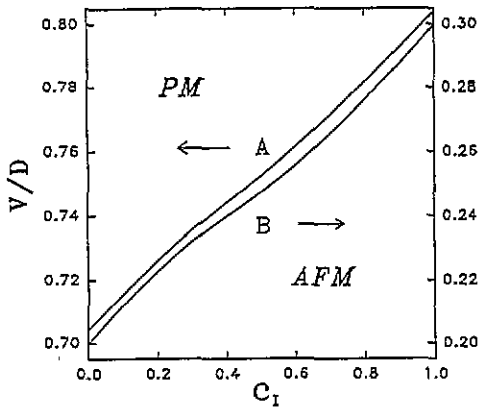

Figure 2. The paramagnetic–antiferromagnetic phase diagram in the c_I–V/D plane with $\Delta V/D = -0.1$; phase boundary A: $n_c = 1$; phase boundary B: $n_c = 0.7$.

It is this equation that determines the boundary of antiferromagnetic instability of HF alloys.

The determination of the phase boundary involves the solution of a system of equations composed of the phase-boundary equation (51) and saddle-point equations (37)–(40), (43) and (44). In these equations, the integral of the imaginary part of $\mathcal{F}_0$ and $\partial\mathcal{F}_I/\partial z_I(\lambda_I^{(2)})$ must be calculated by solving the CPA equations (32) and (34)–(36) self-consistently. The numerical results are shown in figures 1 and 2. In the calculation, we take $E_0/D = -0.8$ and $U \to \infty$. From figure 1, it can be seen that the CB doping, which may decrease or increase the c–f mixing strength V, will lead to expansion or contraction of the region of antiferromagnetism, and the larger the c-electron concentration n_c, the more instable against antiferromagnetism the system becomes. In figure 2, the critical value of the c–f mixing V with constant n_c and negative ΔV is shown as an increasing function of the CB impurity concentration c_I. Obviously, the alloying tends to form an antiferromagnetic state with n_c unchanged if the CB impurities result in a suppression of c–f mixing. The results shown in figure 2 provide a qualitative explanation for the magnetic order displayed in realistic materials such as $Ce(Cu_{1-x}Ag_x)_6$ and $Ce(Cu_{1-x}Au_x)_6$. Since $CeCu_6$ totters on the very edge of antiferromagnetism, a fairly dilute doping with Ag or Au can lead to a paramagnetic–antiferromagnetic phase transition, as observed experimentally [7–9].

5. Summary

In this paper we have presented the results of some theoretical research on the magnetic instability of HF alloys with CB impurities via calculation of the paramagnetic susceptibility. First, a SBMF model including the impurities was established by introducing an infinitesimal staggered magnetic field in the paramagnetic state. Then, within the single-site CPA, the effect of the impurity was treated in two parts: a magnetic-field-independent part and a staggered part proportional to the field. By setting the susceptibility to be divergent, the paramagnetic–antiferromagnetic phase boundary was calculated. Our results show that the IED inducing the increment of cell volume at CB impurity sites favours the formation of an antiferromagnetic state, which agrees with the alloying-induced paramagnetic–antiferromagnetic phase transition detected in $CeCu_6$ [7–9].

Our theory can also be used to explain the effect of non-isoelectronic doping, as in $Ce(Cu_{1-x}Al_x)_6$ [6]. Provided that the c_I dependence of the c-electron concentration n_c has been given, the variation of the paramagnetic–antiferromagnetic phase boundary with the impurity concentration can be directly obtained along the given c_I–n_c line of the phase diagram in the n_c–V/D plane. In a real HF alloy such as $Ce(Cu_{1-x}Al_x)_6$, the contribution of the reduction of cell volume to the formation of a non-magnetic state dominates in comparison with the contribution of the increase of the c-electron concentration to the magnetism, and the system moves away from magnetic order.

Finally, it should be mentioned that in our calculation we assumed a constant variation in c–f mixing strength and neglected multiple scattering among impurities. These approximations do not qualitatively modify the above results.

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